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Engineering ferroelectric domain patterns with ultrafast light

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Ferroelectric domain patterns are essential for variety of applications including quasi-phase matched frequency conversion and beam shaping, high-density non-volatile data storage and memories, photovoltaics, quantum information processing etc. [1]. Typically the electric field poling with patterned electrodes is used to produce ferroelectric domain patterns. However, this technique suffers from low resolution and crystallographic restriction – it is predominantly used in only one crystallographic orientation (the z-cut crystals). The latter aspect is particularly critical since many devices require other crystallographic orientations, e.g. surface sound wave modulator in Lithium Niobate, which requires the x-cut crystal. To overcome these drawbacks all-optical poling was explored [2]. This method uses UV light whose absorption induces high temperature gradient and subsequent domain reversal via thermoelectric field. Due to inherent strong absorption the UV poling is effective in surface layer only. Moreover, surface damage often accompanies domain reversal such that it cannot be applied to waveguides or thin films.

We have recently achieved a breakthrough in ferroelectric domain patterning in lithium niobate crystal by employing femtosecond, infrared laser pulses [3,4]. The method relies on nonlinear absorption of light in beam's focal volume, which induces high temperature gradient and appearance of thermal electric field that locally inverts direction of spontaneous polarization [Figs. 1(a-c)]. The technique enables formation of long domains extending from surface up to 100 microns deep in the crystal [Fig. 1(d)], is applicable to different crystallographic orientation [Fig. 1(d)], and offers a good spatial resolution (~1.0 μm). Moreover, this approach does not induce any damage to the surface and hence can be used with waveguide and membranes of ferroelectric crystals. As an example we applied it to fabricate periodic quasi-phase matching structure in a lithium niobate channel waveguide and achieved efficient SHG with 17.5% conversion efficiency [Figs. 1(e-g)]. Finally, as the ultrafast poling takes place in focal volume of the beam, it opens up possibility to realize true 3-dimensional domain patterning in the bulk of ferroelectric crystal.

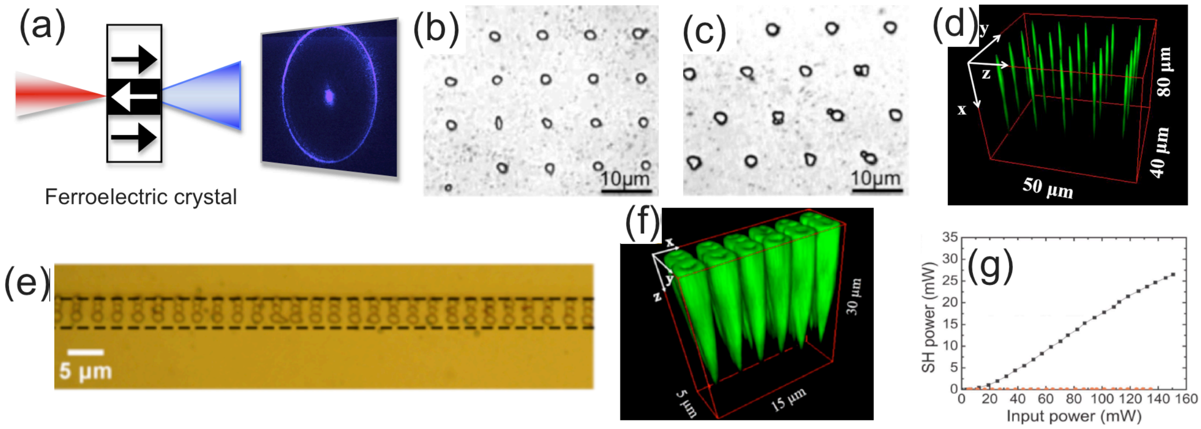

Figure 1 (a) Illustrating principle of light-induced ferroelectric domain reversal. Tightly focused ultrafast beam creates local antiparallel ferroelectric domain in initially homogeneous crystal. The domain reversal is indicated by appearance of the conical Cerenkov second harmonic signal, which provides unique real-time monitoring of the process; (b) and (c) Optical microscopic images of the resulting two-dimensional ferroelectric domain patterns (after HF etching) in z-cut lithium niobate; (d) 3D visualization of a section of square pattern of optically inverted domains in x-cut lithium niobate; (e) and (f) The optical and Cerenkov second harmonic microscopic images of the all-optically poled domain pattern in channel waveguide in z-cut lithium niobate; (g) The measured power of the generated second harmonic vs. the power of the fundamental wave.

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